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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : C08F 220/34, C11D 3/37	A1	(11) International Publication Number: WO 99/06455 (43) International Publication Date: 11 February 1999 (11.02.99)
(21) International Application Number: PCT/EP98/04611 (22) International Filing Date: 22 July 1998 (22.07.98) (30) Priority Data: MI97A001824 30 July 1997 (30.07.97) IT (71) Applicant (for all designated States except US): 3V SIGMA S.P.A. [IT/IT]; Piazzale Principessa Clotilde, 6, I-20121 Milano (IT). (72) Inventors; and (75) Inventors/Applicants (for US only): BERTE', Ferruccio [IT/IT]; Via Torquato Tasso, 58, I-24100 Bergamo (IT). POLOTTI, Gianmarco [IT/IT]; Via Torquato Tasso, 58, I-24100 Bergamo (IT). (74) Agent: MINOJA, Fabrizio; Bianchetti Bracco Minoja S.r.l., Via Rossini, 8, I-20122 Milano (IT).		(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, HR, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: THICKENING AGENTS FOR ACIDIC AQUEOUS COMPOSITIONS <div style="text-align: center;"> $\begin{array}{c} \text{R} \quad \text{O} \quad \text{R}_3 \\ \quad \quad \\ \text{R}_1-\text{CH}=\text{C}-\text{C}-\text{O}-\text{R}_2-\text{N}^+-\text{R}_4 \quad \text{X}^- \\ \\ \text{R}_5 \end{array} \quad (I)$ </div> (57) Abstract <p>The use of cross-linked cationic homopolymers as thickening agents for acidic aqueous compositions, for example detergents for toilet cleaning and laundry softeners. The cross-linked cationic homopolymers are homopolymers of cationic monomers of formula (I) cross-linked with a cross-linking agent present in amounts ranging from not less than 50 to about 600 ppm of the homopolymer total weight. Acidic aqueous compositions thickened with said cross-linked cationic homopolymers. The above mentioned cross-linked cationic homopolymers with a cross-linking agent present in amounts ranging from not less than 50 to about 600 ppm of the homopolymer total weight.</p>		

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THICKENING AGENTS FOR ACIDIC AQUEOUS COMPOSITIONS

The present invention relates to the use of specific crosslinked cationic homopolymers as thickening agents for acidic aqueous compositions. Typical acidic aqueous compositions which can be thickened by the crosslinked cationic homopolymers described in the invention are those for domestic use, such as toilet cleaners and laundry softeners.

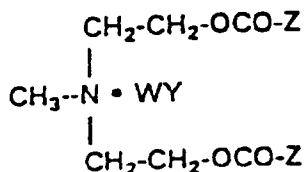
The first ones comprise, as active ingredients, acids or mixtures of acids such as hydrochloric, phosphoric, maleic, citric, oxalic, malonic, sulfamic acid and analogs, whereas laundry softeners are compositions which comprise cationic surfactants as active ingredients. Examples of these surfactants are long alkyl chain dialkyldimethylammonium salts which are, however, used in limited amounts due to their biodegradability problems. In fact, they have been replaced, partly or completely, by cationic surfactants belonging to the family of the so-called "ester quats" (G.R. Whalley, *Happi*, February 1995, page 55; I. Shapiro et al., *Cosmetics and Toiletries*, 109, 77, 1994), which are quaternary ammonium salts, containing at least a group $Z-CO-O-$, in which Z is a straight or branched (C_{8-25}) alkyl, saturated or unsaturated, characterized in that the non-carbonylic oxygen of such group is linked to the quaternary nitrogen through a straight or branched (C_{1-5}), preferably (C_{2-3}), alkylene chain.

Typical non-limiting examples of "ester quats" are
A) imidazole derivatives of formula

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$$\begin{array}{c} \text{Z-CO-O} \\ \text{Z-CO-O-CH}_2 \end{array} \text{CH-CH}_2\text{-N(CH}_3)_2 \bullet \text{WY}$$
$$\text{Z}-\text{CO}-\text{O}-\text{CH}_2-\text{CH}_2-\overset{\text{WY}}{\underset{\bullet}{\text{N}}}-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{NH}-\text{CO}-\text{Z}$$

15



The acidic aqueous compositions which are referred to in the present invention often contain other auxiliary components, such as perfumes, dyes, optical bleachers and analogs, and are in the form of very fluid aqueous systems, i.e. with viscosity values close to those of water. For this reason, they suffer from problems which make their use awkward, such as the outflow of the liquid from the container in an undesired

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amount, with the consequent use of excessive amounts of the composition which, as in the case of laundry softeners, can, although temporarily, affect the properties of the fabric, or involve repeated rinses, with a waste of water, time and power. A further problem, mainly connected with marketing reasons, is that compositions with fluidity similar to water lack that "creamy" consistence that better gives the idea of softness.

On the other hand, when very sloped or even vertical ceramic surfaces are to be cleaned, as is the case of tiled walls, wash-basins or water-closets, the fluidity of the composition makes it to quickly flow over said surfaces, thus decreasing the contact times between the active components of the composition and soil, without attaining its homogeneous distribution on them, so that remarkable amounts of the composition itself are necessary to obtain the desired effect.

Conventional thickeners have been used, such as cellulose derivatives, guar gum, xanthane gum, water-soluble anionic polymers either non crosslinked or partially crosslinked, with unsatisfactory results.

It is anyway common practice to thicken aqueous compositions by adding synthetic polymer compounds, whose nature depends on the intended use of the thickened composition, in order to overcome this and other drawbacks. The choice of the suitable thickening agent depends on the type of the composition and on its use. Anionic or cationic polymeric thickeners can be used. Very frequently, they are crosslinked copolymers of acrylamide and other ethylenically unsaturated

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monomeric units, in which the crosslinking agent is present in amounts which depend on the desired crosslinking degree and, as a consequence, on the intended use of the copolymer (see US 3 968 037 and US 4 806 345). EP 395 282 suggests the use of crosslinked cationic homopolymers and copolymers to thicken particularly acidic aqueous systems ($\text{pH} < 4$), for example solutions for cleaning metals, for removing rust, germicides, detergents for ceramic articles such as those based on citric, phosphoric and hydrochloric acids, in which the crosslinking agent has to be present in very specific amounts (from 5 to no more than 45 ppm - parts per million) of the polymer weight, in that the best performances are reportedly attained only within such range of crosslinking agent. In practice, however, EP 395 282 discloses the use of acrylamide/dimethylaminoethyl methacrylate copolymers quaternised with CH_3Cl . Finally, EP 494 554 describes cationic acrylamide/dimethylaminoethyl methacrylate copolymers quaternised with CH_3Cl , in which the weight ratio of the two monomers is preferably 80 : 20, crosslinked with 50 to 100 ppm of bisacrylamidoacetic acid. These crosslinked cationic copolymers are used as thickening agents in laundry softeners for domestic use. However, acrylamide, which is still present in the final products, is known to be a highly toxic compound. Therefore, a thickening agent for acidic aqueous compositions whose preparation does not involve the use of said substance, is highly requested also from the industrial point of view.

As stated above, the present invention relates to

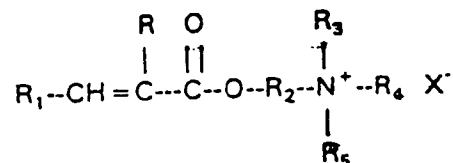
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the use of specific crosslinked cationic homopolymers as thickening agents for acidic aqueous compositions. More particularly, said crosslinked cationic homopolymers are homopolymers of cationic monomers of formula I

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crosslinked with a crosslinking agent present in amounts ranging from not less than 50 to about 600 ppm of the homopolymer total weight.

A further object of the present invention relates to the use of the above mentioned crosslinked cationic homopolymers as thickening agents for acidic aqueous compositions containing an "ester quat" cationic surfactant.

In formula I, R is hydrogen, (C₁₋₄)-alkyl, or the radical CH₂COOH, R₁ is hydrogen, methyl, carboxy, or the radical CH₃--CH=CH, R₂ is methylene or a straight or branched (C₂₋₄) alkylene moiety, R₃, R₄ and R₅ are independently hydrogen or (C₁₋₄) alkyl and X is chlorine, bromine, iodine, hydrogen sulfate or methosulfate.

(C₁₋₄)Alkyl substantially means methyl, ethyl, propyl, isopropyl, n.-butyl, isobutyl, sec.-butyl and tert.-butyl, whereas a straight or branched (C₂₋₄) alkylene moiety substantially means ethylene, 1,3-propylene, 1- and 2-methyl-ethylene, 1,4-butylene, isobutylene and analogs.

A preferred group of compounds of formula I are those in which R is hydrogen or methyl, R₁ is hydrogen

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or methyl, R_2 is methylene, ethylene or 1,3-propylene, R_3 , R_4 and R_5 are independently hydrogen, methyl or ethyl and X is chlorine, bromine, hydrogen sulfate and methosulfate.

5 A second preferred group of compounds of formula I are those in which R is hydrogen or methyl, R_1 is hydrogen, R_2 is methylene or ethylene, R_3 , R_4 and R_5 are methyl and X is chlorine, hydrogen sulfate and methosulfate.

10 All the compounds containing at least two ethylenically unsaturated moieties can be used as crosslinking agents. Illustrative examples of said crosslinking agents are divinyl benzene, allyl acrylates and methacrylates, diacrylates and dimethacrylates of
15 glycols and polyglycols, butadiene, 1,7-octadiene, allyl-acrylamides and allyl-methacrylamides, bisacrylamidoacetic acid, N,N'-methylene-bisacrylamide and polyol polyallylethers, such as polyallylsaccharose and pentaerythritol triallylether. Preferred
20 crosslinking agents are allyl-acrylamides and allyl-methacrylamides, bisacrylamidoacetic acid and N,N'-methylene-bisacrylamide. The most preferred crosslinking agent is N,N'-methylene-bisacrylamide.

The crosslinking agents are used in amounts ranging
25 from not less than 50 ppm to about 600 ppm of the homopolymer total weight. Preferably, the crosslinking agent is present in amounts from about 60 to about 250 ppm of the homopolymer total weight. Contrary to what could be expected, it has surprisingly been found that
30 the viscosities of the acidic aqueous compositions remained substantially steady with increasing amounts of

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crosslinking agent, while the rheology properties of the compositions improved. From the practical point of view, the thus thickened acidic aqueous compositions are easy to handle and to use, they flow out from the container only in the desired amounts and are devoid of substantial stringy consistency. Moreover, the thus thickened acidic aqueous compositions proved to be stable to storage, as substantially neither sedimentation products nor appreciable changes in viscosity are observed, even after long times.

The thickening agents used for the purposes of the present invention are added to acidic aqueous compositions in amounts ranging from about 0.01 to about 5% of the weight of the compositions themselves. Preferably, the amounts used vary from about 0.1 to about 3.5% of the composition weight.

Therefore, a further object of the present invention comprises acidic aqueous compositions thickened with from about 0.01 to about 5% by weight of one or more of the thickening agents described in the present invention and, preferably, with from about 0.1 to about 3.5% of the composition weight.

A further object of the present invention relates to acidic aqueous compositions comprising an "ester quat" cationic surfactant, thickened with from about 0.01 to about 5% by weight of one or more of the thickening agents described in the present invention and, preferably, with from about 0.1 to about 3.5% of the composition weight.

The crosslinked cationic homopolymers used as thickening agents for acidic aqueous compositions,

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according to what described in the present invention,
are products partly known in literature, including the
patent one (see, for instance, US 4,172,066).
Nevertheless, crosslinked cationic homopolymers of
5 monomeric units of formula I in which the crosslinking
agent is N,N'-methylene-bisacrylamide, present in
amounts ranging from not less than 50 to about 600 ppm
of polymer total weight, preferably from about
100 to about 250 ppm, have never expressly been
1 described. Therefore, a further object of the present
invention relates to the above mentioned crosslinked
cationic homopolymers.

Crosslinked cationic homopolymers used as
thickening agents for acidic aqueous compositions,
15 according to what described in the present invention,
can be prepared according to the conventional
polymerization techniques. Reverse emulsion
polymerization is preferred in which, for example, an
aqueous solution of the selected monomer and of the
20 crosslinking agent is emulsified in a phase consisting
of one or more water-immiscible organic liquids. The
water-immiscible organic liquids are, generally, oily
liquids, such as straight or branched (C₅₋₂₀) saturated
hydrocarbons, vaseline oil, aromatic hydrocarbons such
25 as benzene, toluene and xylenes, halo solvents, or
mixtures thereof. The oily phase can contain an
emulsifying system, typically one having an HLB
(Hydrophilic-Lipophilic Balance) ranging from about 2 to
about 7, comprising, for example, sorbitan esters,
30 glycerol esters and analogs or mixtures thereof, to
promote the dispersion of the aqueous phase which, in

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addition to the monomer of formula I and the crosslinking agent in the selected amounts, also contains one of the polymerization initiators commonly used in the art, such as the product marketed as WAKO^(R) V 50. The polymerization is carried out at temperatures ranging from about room temperature to about 100°C, and is completed in substantially quantitative yields in a time which can range from about 1 to about 12 hours. The desired crosslinked cationic homopolymer can be recovered in the solid form, if desired, by distillation of the reaction mixture under vacuum. Said solid form is then ground and sieved to obtain a powder which is easily dispersible in the acidic aqueous composition. Alternatively, a homogeneous dispersion of the polymer in the oily phase, both in the hydrated and in the anhydrous forms, can be obtained by azeotropical distillation of water. In this case, it is preferable to add, at the end of the process, a given amount of a phase inversion surfactant with HLB ranging from about 8 to about 16, such as ethoxylated lauryl alcohol with ethoxylation degree from 4 to 12, which does not affect the stability of the emulsion but quickens the dispersion of the emulsion when added to the acidic aqueous compositions. This oily homogeneous dispersion can be added as it is to the acidic aqueous composition. For the purposes of the present invention, the use of the crosslinked cationic homopolymers in the form of solid dispersible powders is preferred.

The preparation of the thickened acidic aqueous composition is also carried out conventionally, adding the fluid composition with the given amount of

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crosslinked cationic homopolymer, or with a mixture of said homopolymers, either as dispersible solid powders or in the form of oily homogeneous dispersions, so as to obtain a composition having the desired viscosity and rheology.

The following examples further illustrate the invention.

Preparation of crosslinked cationic homopolymers as powders

10 A solution A is prepared, consisting of 438.0 g of a 75% solution of dimethylaminoethyl methacrylate quaternised with CH_3Cl in demineralized water, x grams of a 1% solution of N,N'-methylene-bisacrylamide (the values of x will be reported in table 1 below) in demineralized water, (20-x) grams of demineralized water, 0.1 g of DISSOLVINE^(R) D-40 and 1.5 g of 10% WAKO^(R) V-50 in demineralized water. Solution B is then prepared, consisting of 150 g of n-decane and 15 g of SPAN^(R) 80.

20 Solution A is emulsified in solution B. The resulting emulsion is poured into a polymerization reactor and, after removing oxygen by nitrogen bubbling, the polymerization is carried on at 60°C for 6 hours. Upon distillation under vacuum at temperatures comprised between 20 and 120°C, a solid is recovered, which is ground and sieved (hole diameter: 1 - 2 mm), to obtain an easy-to-use powder which is easily dispersible in an aqueous medium.

30 Table 1 reports the amounts x of 1% solution of N,N'-methylene-bisacrylamide in demineralized water and its respective amounts expressed in ppm. The table also

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reports the viscosities of two commercially available
laundry softeners [LENOR^(R), PROCTER & GAMBLE Italia
S.p.A. (comp. A) and COCCOLINO^(R), UNILEVER Italia
S.p.A. (comp. B), both comprising "ester quat" cationic
5 surfactants] thickened with 0.3% by weight of the
crosslinked cationic homopolymers with the given amounts
of N,N'-methylene-bisacrylamide (NN-MBA), prepared as
described above. Viscosities were measured with a
Brookfield RVT viscosimetre (T. 20°C, 20 rpm, spindler
10 5) after the compositions reached their equilibrium, and
are expressed in cps (centipoises).

TABLE 1

15 Ex.	x (g)	NN-MBA (ppm)	Viscosity (comp. A) (cps)	Viscosity (comp. B) (cps).
1	2	60	1000	1500
2	2.3	70	1200	1500
20 3	3	90	1000	1500
4	4	120	1000	1400

The thickened compositions did not substantially
show stringy character. Viscosity and rheology remained
25 unchanged up to 5 weeks. Analogous results were obtained
thickening 5% aqueous solutions of "ester quat" cationic
surfactants.

Table 2 reports the viscosity of a commercially
available strongly acidic detergent composition for
30 removing lime scales from ceramic surfaces (VIAKAL^(R),
PROCTER & GAMBLE Italia S.p.A.), thickened with 2.5% by
weight of the cationic homopolymers crosslinked with the

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given amounts of N,N'-methylene-bisacrylamide (NN-MBA), prepared as described above. The meaning of x is the same as that in table 1. The viscosities were measured and expressed as indicated above.

5 TABLE 2

Ex.	x (g)	NN-MBA (ppm)	Viscosity (cps)
10 5	2	60	2500
6	2.3	70	3000
7	3	90	1800

15 The thickened compositions did not substantially show stringy character. Viscosity and rheology remained unchanged for up to 5 weeks.

Preparation of crosslinked cationic homopolymers as emulsions

20 A solution A is prepared, consisting of 438.0 g of a 75% solution of dimethylaminoethyl methacrylate quaternised with CH₃Cl in demineralized water, x grams of a 1% solution of N,N'-methylene-bisacrylamide (the values of x will be reported in table 3 below) in demineralized water, (20-x) grams of demineralized
25 water, 0.1 g of DISSOLVINE^(R) D-40 and 1.5 g of 10% WAKO^(R) V-50 in demineralized water. A solution B is then prepared, consisting of 250.0 g of vaseline oil, 75.0 g of n-decane and 15.0 g of SPAN^(R) 80. Solution A is emulsified in solution B. The resulting emulsion is
30 poured into a polymerization reactor and, after removing oxygen by nitrogen bubbling, the polymerization is

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carried on at 60°C for 6 hours. First, water is
distilled off azeotropically between 90 and 110°C, then
n-decane is also distilled. The mixture is cooled at
room temperature, and subsequently added with 20.0 g of
5 ethoxylated lauryl alcohol with ethoxylation degree 7,
thus obtaining a homogeneous, stable dispersion of the
crosslinked cationic homopolymer in vaseline oil, having
a dull white appearance and a viscosity of 1800 cps
(centipoises), measured with a Brookfield RVT
10 viscosimetre, spindler 3, at 20°C and 20 rpm. The
resulting product is used directly to thicken a laundry
softener composition.

Table 3 reports the amounts x of 1% solution of
N,N'-methylene-bisacrylamide in demineralized water and
15 the respective amounts of NN-MBA expressed in ppm. The
table also reports the viscosity of a commercially
available laundry softener (COCCOLINO^(R), UNILEVER
Italia S.p.A.) thickened with 0.5% by weight of the
homogeneous dispersion in vaseline oil prepared as
20 described above. Said amount corresponds to about 0.25%
of crosslinked cationic homopolymer with the givens
amounts of N,N'-methylene-bisacrylamide (NN-MBA).
Viscosities were measured as described above.

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TABLE 3

Ex.	x (g)	NN-MBA (ppm)	Viscosity (cps)
5			
8	2.3	60	1800
9	3	90	2000
10	6	180	1900

10 The thickened compositions did not substantially show stringy character. Viscosity and rheology remained unchanged up to 5 weeks.

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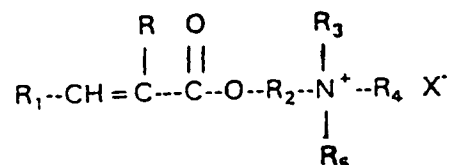
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CLAIMS

1. The use of crosslinked cationic homopolymers of cationic monomers of formula I

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in which:

10 R is hydrogen, (C₁₋₄)alkyl, or the radical CH₂COOH, R₁ is hydrogen, methyl, carboxy or the radical CH₃--CH=CH, R₂ is methylene or a straight or branched (C₂₋₄) alkylene moiety, R₃, R₄ and R₅ are independently hydrogen or (C₁₋₄) alkyl and X is chlorine, bromine, iodine, hydrogen sulfate or methosulfate, said homopolymers being characterised in that they are crosslinked with a crosslinking agent present in amounts ranging from not less than 50 to about 600 ppm of the homopolymer total weight, as thickening agents for acidic aqueous compositions.

20

2. The use of crosslinked cationic homopolymers as claimed in claim 1, in which R is hydrogen or methyl, R₁ is hydrogen or methyl, R₂ is methylene, ethylene or 1,3-propylene, R₃, R₄ and R₅ are independently hydrogen, methyl or ethyl and X is chlorine, bromine, hydrogen sulfate and methosulfate.

25

3. The use of crosslinked cationic homopolymers as claimed in claim 1, in which R is hydrogen or methyl, R₁ is hydrogen, R₂ is methylene or ethylene, R₃, R₄ and R₅ are methyl and X is chlorine, hydrogen sulfate and methosulfate.

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4. The use of crosslinked cationic homopolymers as claimed in any one of claims 1-3, in which the crosslinking agent is selected from allyl-acrylamides, allyl-methacrylamides, bisacrylamidoacetic acid and N,N'-methylene-bisacrylamide.

5. The use of crosslinked cationic homopolymers as claimed in any one of claims 1-3, in which the crosslinking agent is N,N'-methylene-bisacrylamide.

6. The use of crosslinked cationic homopolymers as claimed in any one of claims 1-5, in which the crosslinking agent is present in amounts ranging from about 60 to about 250 ppm of the homopolymer total weight.

7. The use of crosslinked cationic homopolymers as claimed in any one of claims 1-6 in the form of solid dispersible powders.

8. The use of crosslinked cationic homopolymers according to any one of claims 1-7 as thickening agents for acidic aqueous compositions, comprising an "ester quat" cationic surfactant.

9. Acidic aqueous compositions thickened with crosslinked cationic homopolymers according to any one of claims 1 to 7.

10. Acidic aqueous compositions as claimed in claim 9, in which the crosslinked cationic homopolymer used as thickening agent is present in amounts ranging from about 0.01 to about 5% of the composition weight.

11. Acidic aqueous compositions as claimed in claims 9 and 10, in which the crosslinked cationic homopolymer used as thickening agent is present in amounts ranging from about 0.1 to about 3.5% of the composition weight.

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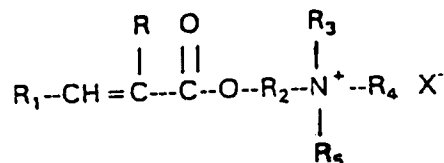
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12. Acidic aqueous compositions as claimed in claims 9 to 11, comprising an "ester quat" cationic surfactant.

13. Crosslinked cationic homopolymers of cationic monomers of formula I

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in which R is hydrogen, (C₁₋₄)-alkyl, or the radical CH₂COOH, R₁ is hydrogen, methyl, carboxy, or the radical CH₃--CH=CH, R₂ is methylene or a straight or branched (C₂₋₄) alkylene moiety, R₃, R₄ and R₅ are independently hydrogen or (C₁₋₄) alkyl and X is chlorine, bromine, iodine, hydrogen sulfate or methosulfate, said homopolymers being characterized in that they are crosslinked with a crosslinking agent present in amounts ranging from not less than 50 to about 600 ppm of the homopolymer total weight.

14. Crosslinked cationic homopolymers as claimed in claim 13 in which R is hydrogen or methyl, R₁ is hydrogen or methyl, R₂ is methylene, ethylene or 1,3-propylene, R₃, R₄ and R₅ are independently hydrogen, methyl or ethyl and X is chlorine, bromine, hydrogen sulfate and methosulfate.

15. Crosslinked cationic homopolymers as claimed in claim 13 in which R is hydrogen or methyl, R₁ is hydrogen, R₂ is methylene or ethylene, R₃, R₄ and R₅ are methyl and X is chlorine, hydrogen sulfate and methosulfate.

16. Crosslinked cationic homopolymers as claimed in any one of claims 13 to 15, in which the crosslinking agent

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is selected from ¹⁸ allyl-acrylamides, allyl-methacrylamides, bisacrylamidoacetic acid and N,N'-methylene-bisacrylamide.

5 17. Crosslinked cationic homopolymers as claimed in any one of claims 13 to 15, in which the crosslinking agent is N,N'-methylene-bisacrylamide.

10 18. Crosslinked cationic homopolymers as claimed in any one of claims 13 to 17 characterized in that the crosslinking agent is present in amounts ranging from about 60 to about 250 ppm of the homopolymer total weight.

INTERNATIONAL SEARCH REPORT

International application No

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A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08F220/34 C11D3/37

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08F C11D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the International search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 0 395 282 A (ALLIED COLLOIDS LTD) 31 October 1990 cited in the application see claims; examples ---	1-18
Y	FR 2 070 686 A (INVERESK RES INT) 17 September 1971 see page 3, line 34 - page 4, line 2; claims; examples ---	1-18
Y	US 4 542 175 A (FINK HERBERT ET AL) 17 September 1985 see claims 1-15; examples 1,3,9-23 see column 2, line 49 - column 3, line 25 -----	1-18

☐ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

25 November 1998

Date of mailing of the international search report

10/12/1998

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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